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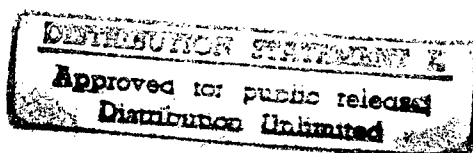
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A PHOTOCHEMICAL METHOD FOR SEPARATING MERCURY ISOTOPES

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A PHOTOCHEMICAL METHOD FOR SEPARATING MERCURY ISOTOPES

[This is a translation of an article written by I. P. Shmelev in Priroda (Nature), No. 1, Moscow, 1955, pages 84-85.]

Isotopes are varieties of a single chemical element having different atomic weights. Thus, for instance, tin (atomic weight 118.70) has 10 isotopes with atomic weights of 112, 114, 115, 116, 117, 118, 119, 120, 122, 124. Because the charge of their atomic nuclei is the same, all of them occupy one and the same place in the periodic table of Mendeleev.

There are many methods of separating individual isotopes. Here, we will examine a method based on the use of photochemical reactions. Photochemical reactions - these are reactions of combinations or dissociations, carried on under the effects of visible or ultraviolet light. Having absorbed a certain portion of the light, atoms or molecules of certain materials achieve excited states and the capacity to enter into chemical reactions, forming molecules of new chemical combination. It is possible to achieve atomic excitation not only with the aid of light, but also by means of high temperatures or electrical discharge.

If atoms that have been excited by some means are left alone, they will revert to their normal state, emitting the excess energy in the form of quanta of electromagnetic radiation with the characteristic frequency of the given atom. As a result, the spectrum emitted by these atoms exhibits the fully defined range of lines present only in that atom. The spectra of the isotopes of one and the same element are absolutely identical in nature. Nevertheless, the wavelengths of corresponding spectral lines of different isotopes (in emission as well as in absorption spectra) differ somewhat from one another. This effect is called the isotopic displacement of spectral lines, or the isotopic effect in atomic spectra. Thanks to isotopic displacement, it is possible to excite the atoms of any given isotope without exciting the others. In this fashion, it is possible to

to formulate the following conditions for the successful application of photochemical reactions to the separation of isotopes of any element:

First, the isotopes of this element or their compounds must be sufficiently different in their absorption of certain wavelengths. Second, it is essential to have a source of light such that, upon the emission of this wavelength, it will excite only the atoms of the desired isotope and not of the others. Third, it is necessary to select a chemical reaction that will take place only in the presence of excited atoms or molecules of the given substance. It is also necessary that conditions be such that the formation of compounds not be followed by their decomposition.

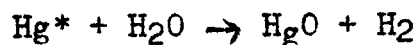
A photochemical method was recently applied to the separation of mercury isotopes.¹ The authors describing the work attempted to concentrate the mercury isotope 198. The spectral lines of this isotope are used in spectroscopy as a standard wavelength. Let us see how the first condition for the spectral lines of the mercury isotopes is fulfilled. The spectral line of mercury chosen lies in the ultraviolet region, and has a wavelength of 2,537 Å (Å - Angstrom - a unit of length, used in spectroscopy, equal to one hundred millionth of a centimeter). The atomic line 2,537 Å consists of several closely separated isotopic lines. The intensity of the line of each isotope depends on the concentration of the isotope in natural mercury. The line representing isotope 198 trails behind the central line by 10.5 Å. Within 15 Å of the line representing isotope 198 are to be found the lines of isotopes 199, 200 and 201. Consequently, upon excitation of the atoms of isotope 198 with a narrow spectral line, it is possible to create weak excitation of these isotopes. The remaining isotopes will not be excited.

How can we obtain a light source of the required wavelength? The light source was a discharge lamp of a type used in medicine and called a quartz lamp. It is filled

¹ See: B.H. Billings, W.J. Hitchcock, and M. Zelikoff, Journal of Chemical Physics, Vol 21, 1953, page 1,762

with the pure, vaporized mercury isotopes 198.²

For the photochemical reaction, that of the oxidation of mercury to HgO was chosen. It was determined that this reaction when carried out in an atmosphere of pure oxygen does not result in any significant concentration of isotope 198 since, under the action of the ultraviolet rays generated by the mercury lamp, ozone is produced in addition to the excitation of Hg¹⁹⁸. Ozone, in turn, forms HgO with the unexcited atoms of the other mercury isotopes. Best results were obtained in the presence of steam. In this case, the reaction is:



Here Hg* represents the excited state of mercury atoms. With nonexcited atoms of mercury practically no reaction takes place, and hence side reactions are almost completely absent.

The apparatus for separating the mercury isotopes was constructed in the following fashion. The reaction took place in a cylindrical vessel in which was placed a discharge tube filled with vaporized Hg¹⁹⁸.

Steam was passed through a layer of liquid mercury and then, with the mercury vapor, passed into the vessel. Emission from the discharge tube preferentially excited the atoms of the mercury isotope 198, which reacted with the molecules of water to form Hg¹⁹⁸O. Then, the steam, the vaporized forms of all the mercury isotopes, and the Hg¹⁹⁸O were collected in a vessel, and the Hg¹⁹⁸O was separated from the mixture.

After a certain period of operation, the vessel was washed with nitric acid thus recovering the oxide of mercury. In the resulting solution a copper wire was introduced, and the concentrated mercury isotope 198 was deposited on the wire forming an amalgam upon its surface. In a special apparatus, the mercury was evaporated from the

² Pure mercury isotope 198 was obtained by the exposure of gold in a nuclear reactor (kettle). Gold (consisting of one isotope with a mass of 197), exposed to neutrons in the kettle, gives a radioactive isotope Au¹⁹⁸, which upon decomposition (half-life of 2.5 hours) turns into mercury 198.

wire, and its vapor led into a discharge tube in which was carried out a spectroscopic analysis of the isotopic contents of the mercury. For the analysis, the spectral line of mercury in the green region of the spectrum (wavelength of 461 Å) was chosen. In the following table are given the results of the spectroscopic analysis before and after concentration.

Isotope	Natural Composition	Natural Concentration
196	very little	very little
198	10%	15%
199	16%	15%
200	24%	26%
201	14%	12%
202	29%	26%
204	7%	6%
	<u>100%</u>	<u>100%</u>

From the table it is seen that the relative amount of isotope Hg^{198} rose to 15%, i.e., increased by one half. This is a very high concentration if it is recalled that the coefficient of concentration of the most widespread industrial methods of separation of isotopes: fractional distillation, diffusion of gases, thermo diffusion, etc., lie within the limits of 1.006-1.1 for a one-stage separation. It may be expected that photochemical means of separating isotopes will turn out to be economical in other cases.

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